ESRF-EBS-NRS Workshop, Grenoble, 11 - 13 March 2019

Sharpening spectral resolution and polarization purity of hard x-rays at the ESRF-EBS

Ralf Röhlsberger

Deutsches Elektronen-Synchrotron DESY, Hamburg

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

DESY.

Outline

(1) Structure and dynamics of complex materials on mesoscopic length scales

(2) Vibrational spectroscopy with μeV resolution

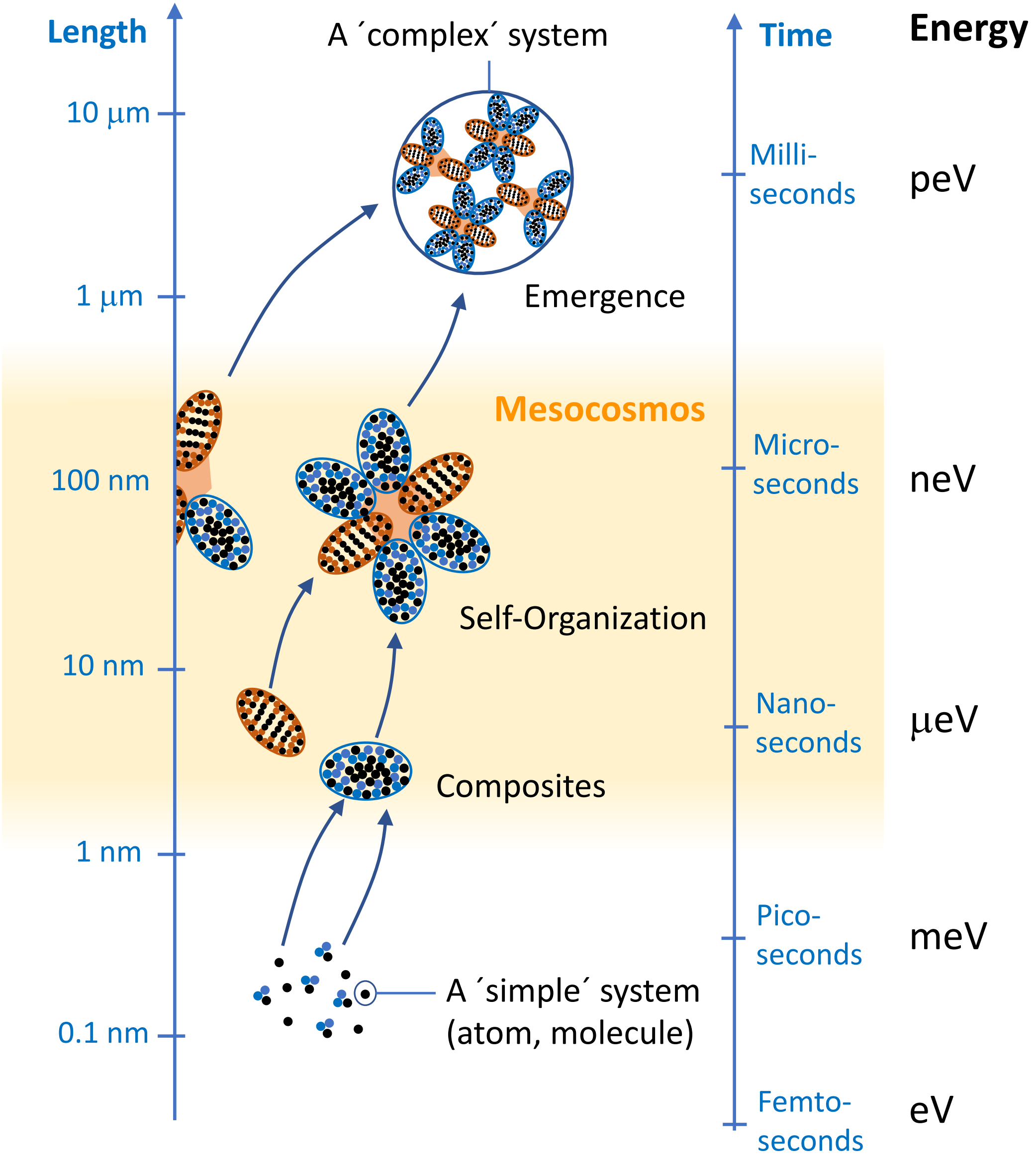
(3) Enabling technology: High purity polarimetry

(4) Applications of high purity polarimetry

- Revealing anisotropies in condensed matter
- Spinwave spectroscopy

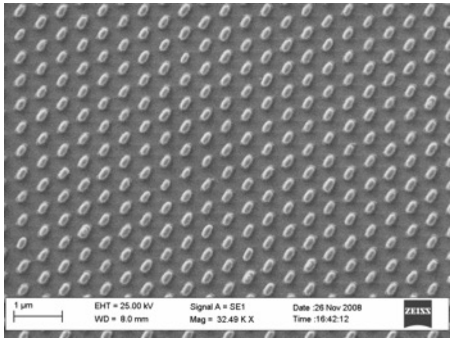
(5) Outlook: High-energy polarimetry

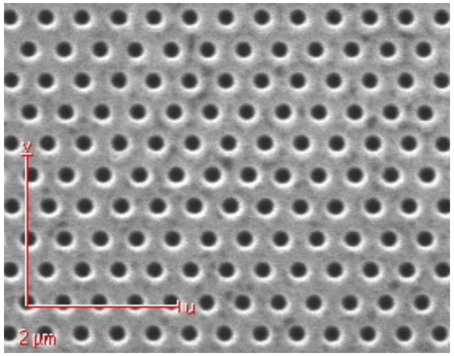
The quest for x-rays with ultrasmall emittance: Understanding dynamics on mesoscopic length scales



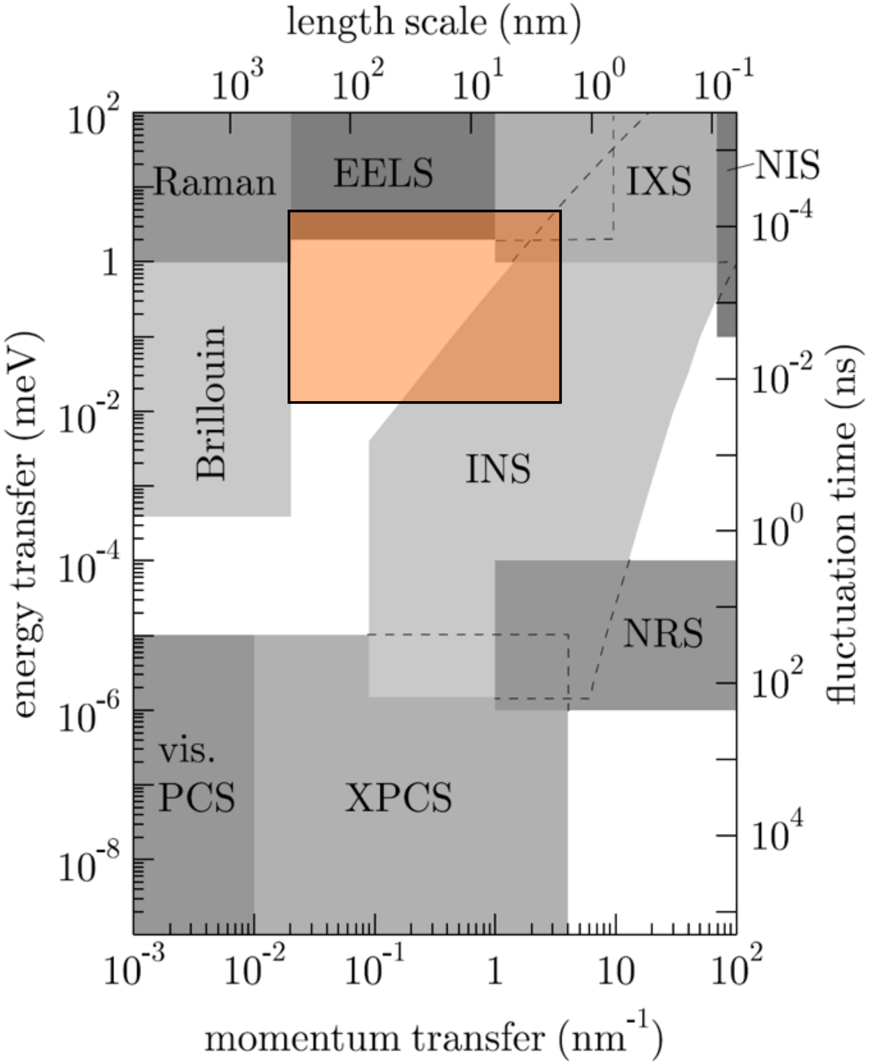
Vibrational dynamics on mesoscopic length scales (1 nm – 100 nm)

Artificially structured materials

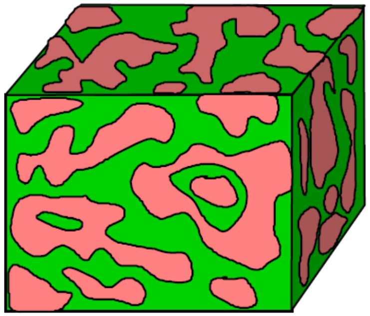


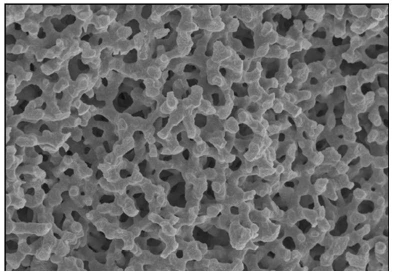


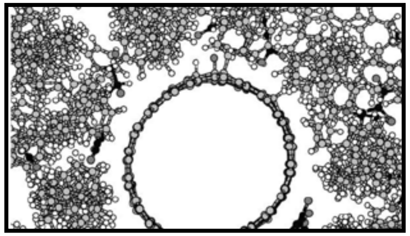
Photonic and phononic crystals



Nanocomposites







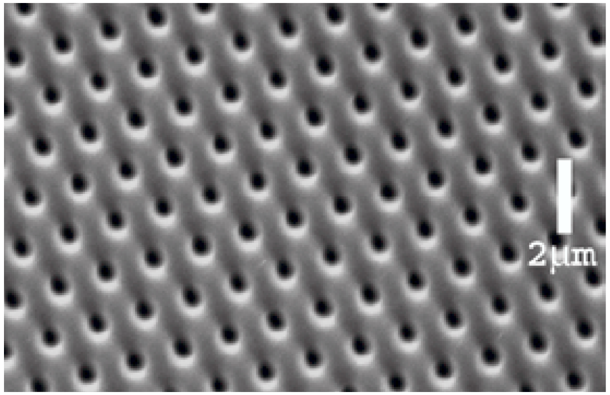
Dynamics of artificially structured materials

Dynamical properties are modified due periodic variation of elastic properties

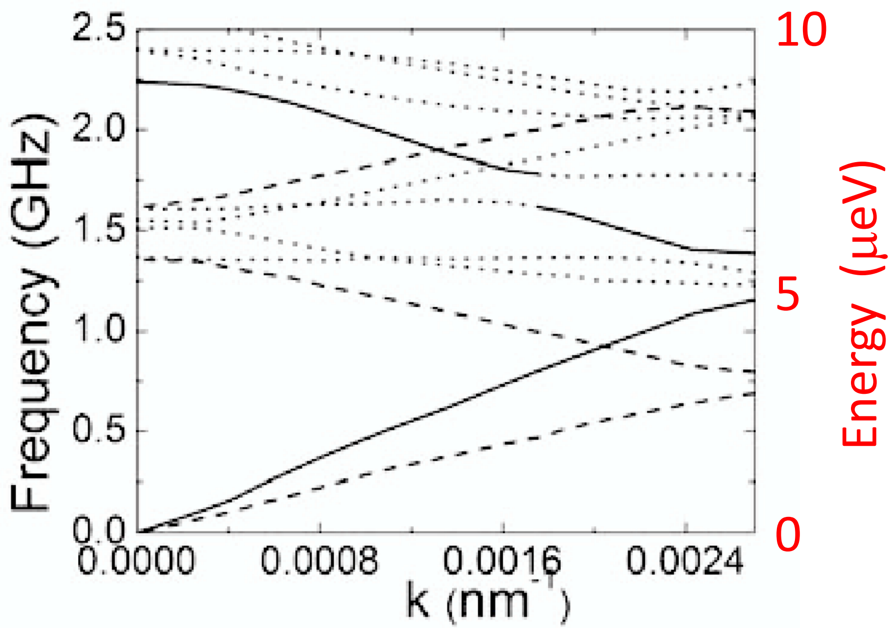
This allows to tailor the vibrational properties of new materials by adjusting their structure

→ Nanocomposites (e.g. metal/polymer, amorphous/crystalline)

Vibrational excitations play an important role for energy dissipation upon friction



Phononic crystal



T. Gorishnyy et al., Phys. Rev. Lett. 94, 115501 (2005)

N·H
ELSEVIER

Nuclear Instruments and Methods in Physics Research A 394 (1997) 251–255

NUCLEAR
INSTRUMENTS
& METHODS
IN PHYSICS
RESEARCH
Section A

X-ray optics for μeV-resolved spectroscopy

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A.I. Chumakov[c], E.E. Alp[a]

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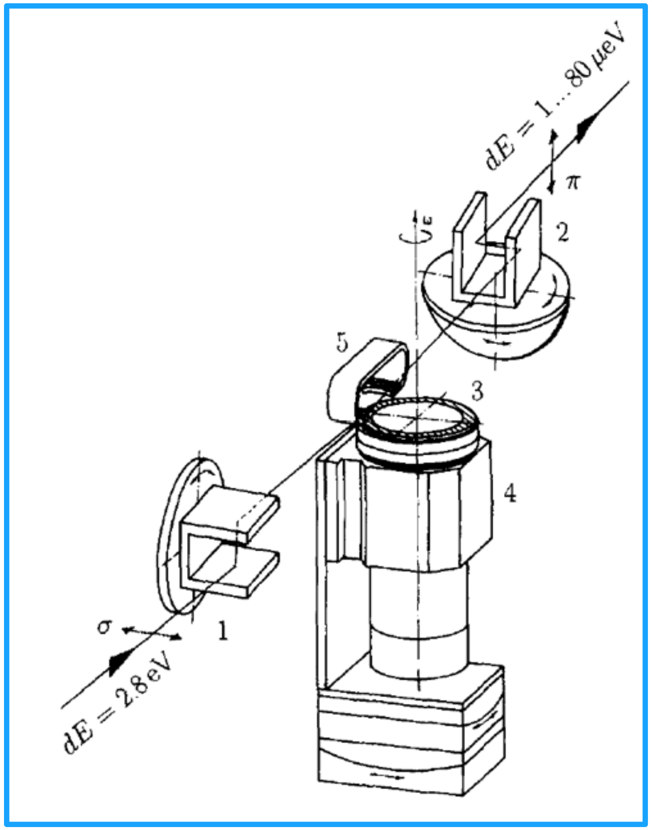
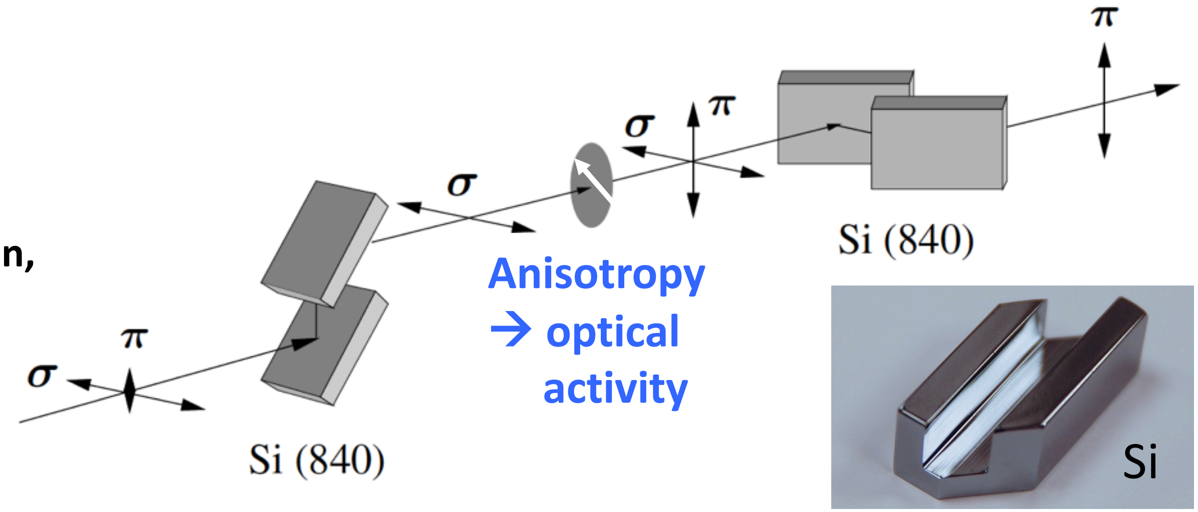
[b] II. Institut für Experimentalphysik, Luruper Chaussee 149, 22761 Hamburg, Germany
[c] European Synchrotron Radiation Facility, BP 220, 38043 Grenoble Cedex, France



Enabling Technology: High-purity Polarimetry

Crossed polarizers for hard X-rays

Collaboration with I. Uschmann, G. Paulus et al., FSU + HI Jena



Anisotropy
→ optical activity

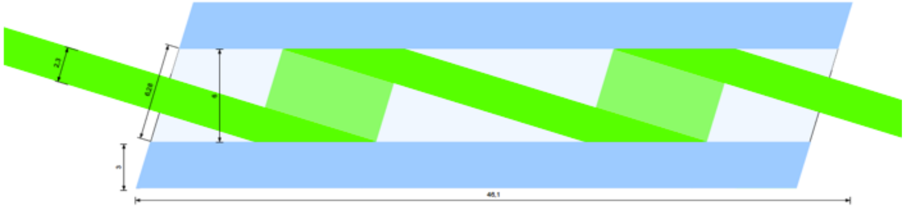
Polarization Purity

$$\delta = I_\pi / I_\sigma = 10^{-10} \dots 10^{-9}$$

- Efficient selection of σ → π scattering
- Detection of very weak anisotropies (linear, circular)

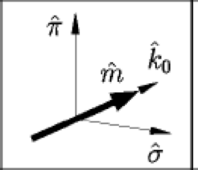
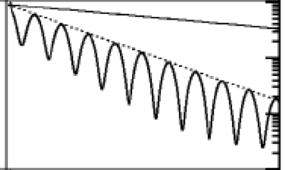
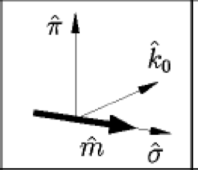
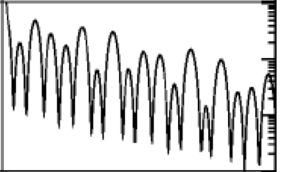
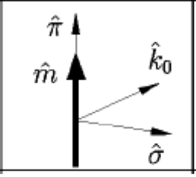
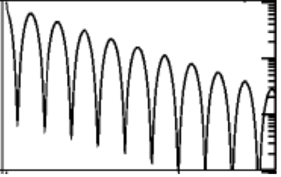
B. Marx et al.,
Opt. Commun. (2011)
Phys. Rev. Lett. (2013)
H. Bernhardt et al.,
Appl. Phys. Lett (2016)

4-bounce polarizer for 14.4 keV (B. Marx et al.)

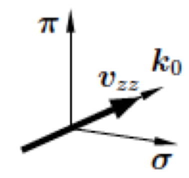
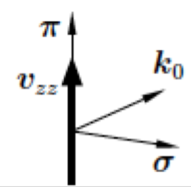
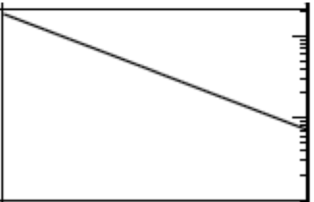
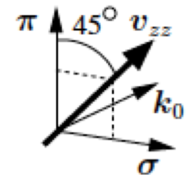


Optical activity in (nuclear) resonant scattering

Magnetic hyperfine interaction

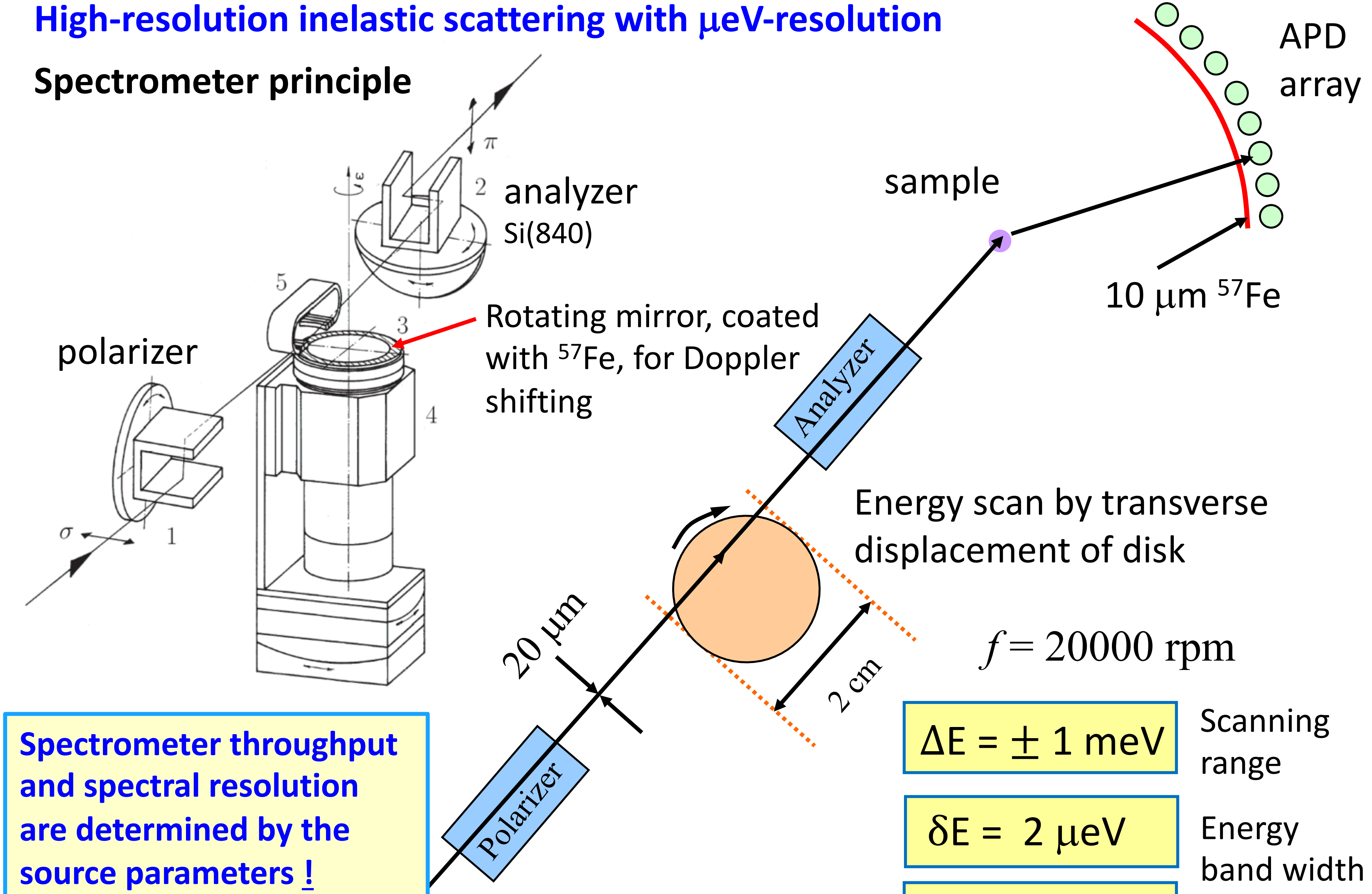
	Geometry	$f_n(\omega)$	Time spectrum $\sigma \to$ unpolarized
A		$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & -\mathrm{i}(F_{+1}-F_{-1}) \\ \mathrm{i}(F_{+1}-F_{-1}) & F_{+1}+F_{-1} \end{pmatrix}$	
B		$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & 2F_0 \end{pmatrix}$	
C		$\frac{3}{16\pi}\begin{pmatrix} 2F_0 & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$	

Electric hyperfine interaction

	Geometry	$f_n(\omega)$	Time spectrum $\sigma \to$ unpolarized
A		$\frac{3}{8\pi}\begin{pmatrix} F_{+1} & 0 \\ 0 & F_{+1} \end{pmatrix}$	
C		$\frac{3}{8\pi}\begin{pmatrix} F_0 & 0 \\ 0 & F_{+1} \end{pmatrix}$	
D		$\frac{3}{16\pi}\begin{pmatrix} F_0+F_{+1} & F_0-F_{+1} \\ F_0-F_{+1} & F_0+F_{+1} \end{pmatrix}$	

High-resolution inelastic scattering with μeV-resolution

Spectrometer principle



polarizer

analyzer
Si(840)

Rotating mirror, coated with ^{57}Fe, for Doppler shifting

sample

APD array

10 μm ^{57}Fe

Energy scan by transverse displacement of disk

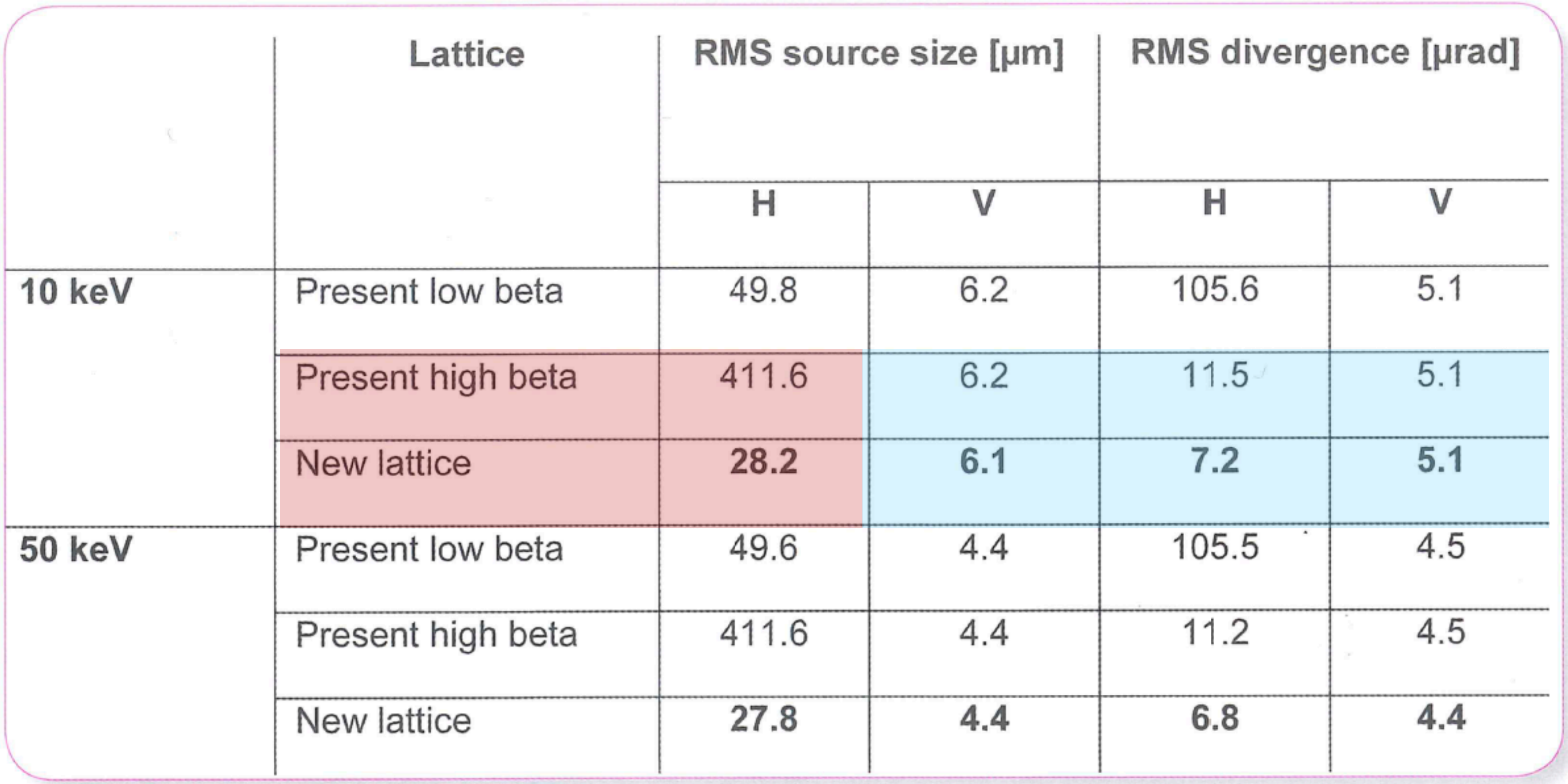
$f = 20000$ rpm

ΔE = ± 1 meV	Scanning range
δE = 2 μeV	Energy band width
F ~ 10^7 ph/s	Photon flux

Spectrometer throughput and spectral resolution are determined by the source parameters !

ESRF-EBS Source Parameters

4 m undulator (18 mm period, k = 2)



	Lattice	RMS source size [μm]		RMS divergence [μrad]	
		H	V	H	V
10 keV	Present low beta	49.8	6.2	105.6	5.1
	Present high beta	411.6	6.2	11.5	5.1
	New lattice	**28.2**	**6.1**	**7.2**	**5.1**
50 keV	Present low beta	49.6	4.4	105.5	4.5
	Present high beta	411.6	4.4	11.2	4.5
	New lattice	**27.8**	**4.4**	**6.8**	**4.4**

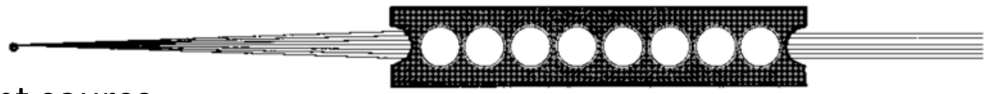
→ Improvements at ID18 mostly due to the small horizontal source size

Polarimetry at ultralow-emittance storage rings

Small source size + collimation

→ small horizontal beam cross section and small divergence

Refractive collimator

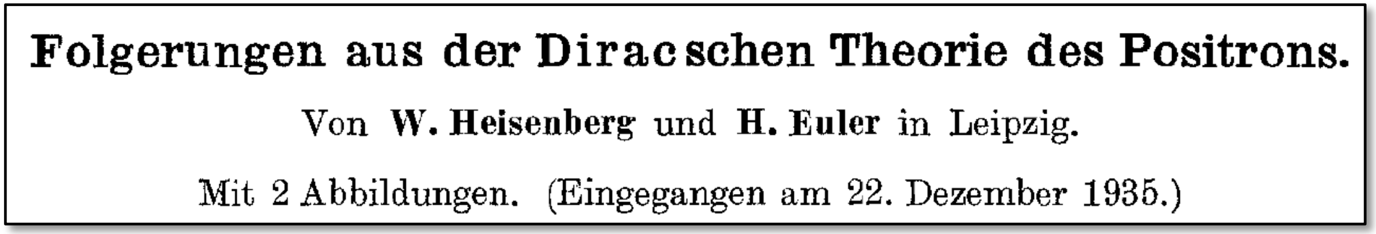


A. Q. R. Baron et al., Appl. Phys. Lett. 74, 1492 (1999)

Collimation down to 1 μrad seems feasible at the ESRF-EBS

Values < 1 μrad in combination with asymmetric Bragg reflections

The quest for ultrahigh purities: Polarization of the Vacuum


Folgerungen aus der Diracschen Theorie des Positrons.

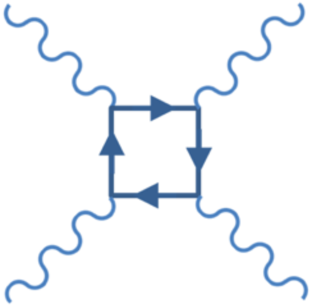
Von **W. Heisenberg** und **H. Euler** in Leipzig.

Mit 2 Abbildungen. (Eingegangen am 22. Dezember 1935.)

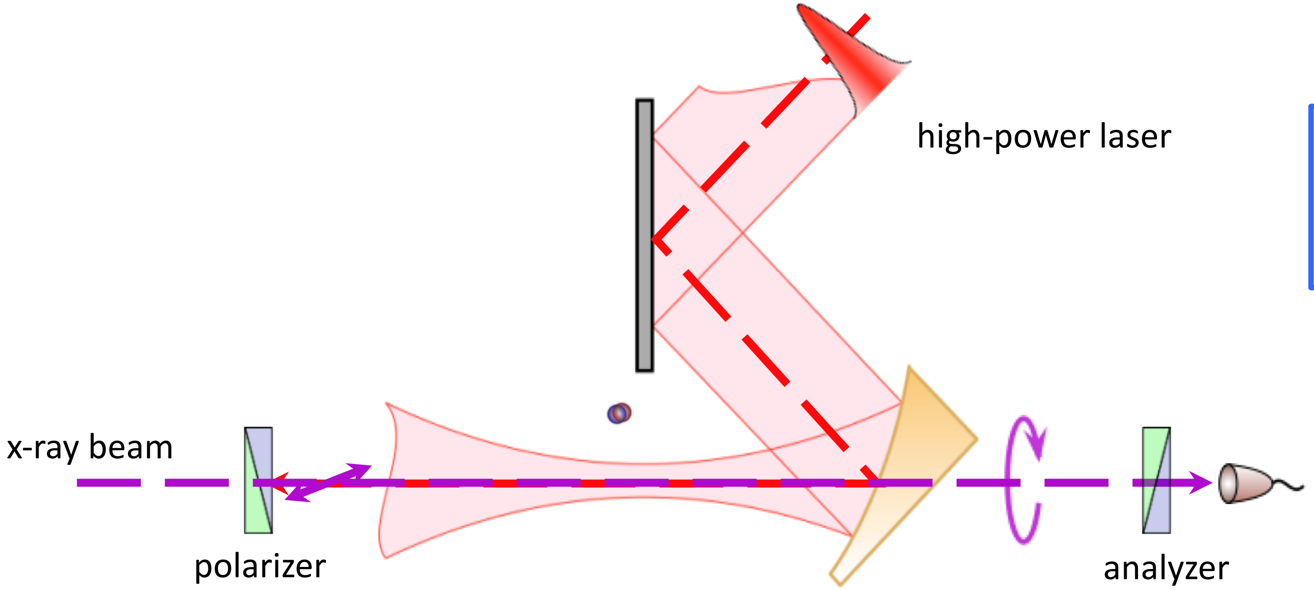
Z. Physik 98, 714 (1936)

„ … even in situations where the photon energy is not sufficient for matter production, its virtual possibility will result in a ´polarization of vacuum´ and hence in an alteration of Maxwell´s equations“

→ vacuum becomes birefringent


Photon-photon scattering

Detection of Vacuum Birefringence: Probing the quantum vacuum



Ellipticity

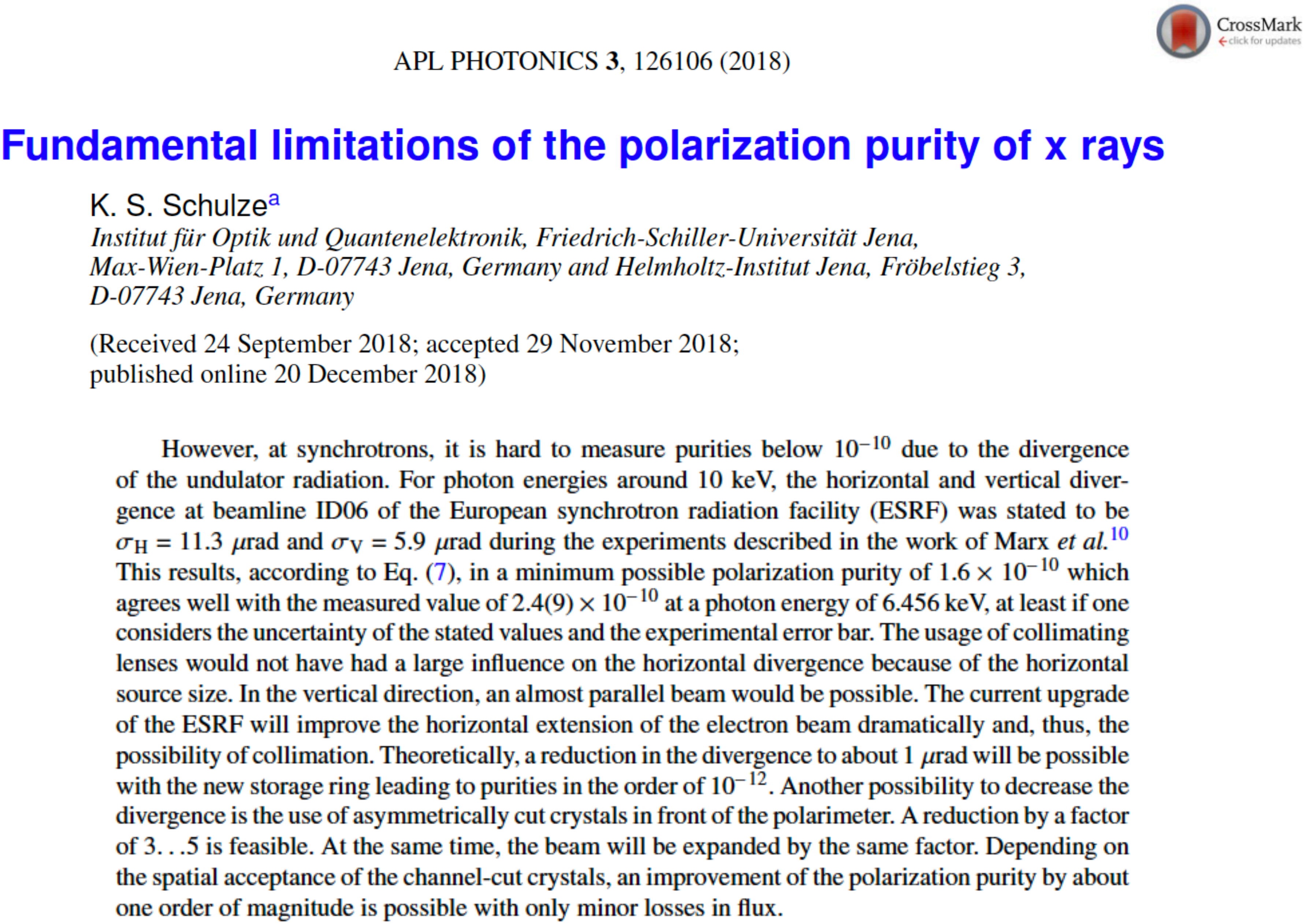
$$\Delta\phi = \frac{4\alpha}{15}\frac{z_0}{\lambda}\frac{I_0}{I_C}$$

α	fine structure constant
z_0	interaction length
λ	probe wavelength
I_0	electric laser field
I_C	critical field for pair production = 4.4 x 10^{29} W/cm²

for $I_0 = 10^{22}$ W/cm² and λ = 1 Å → **ellipticity < 10^{-12} expected**

→ **Purities δ ~ 10^{-12} required**

T. Heinzl, R. Sauerbrey et al., Opt. Commun. 267, 318 (2006)



APL PHOTONICS **3**, 126106 (2018)

Fundamental limitations of the polarization purity of x rays

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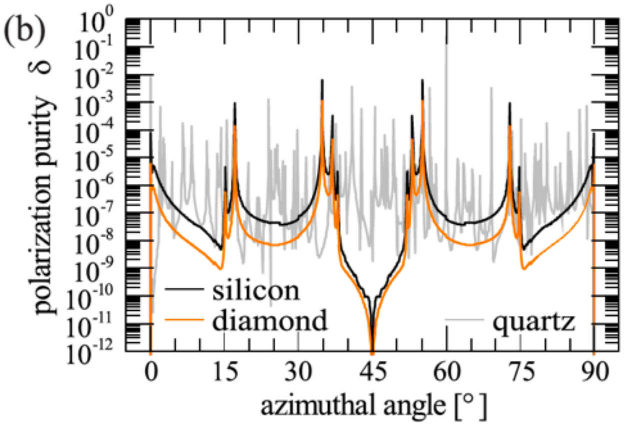
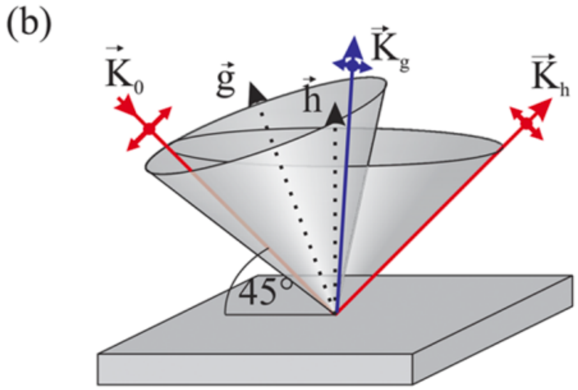
(Received 24 September 2018; accepted 29 November 2018;
published online 20 December 2018)

However, at synchrotrons, it is hard to measure purities below 10^{-10} due to the divergence of the undulator radiation. For photon energies around 10 keV, the horizontal and vertical divergence at beamline ID06 of the European synchrotron radiation facility (ESRF) was stated to be $\sigma_{\mathrm{H}} = 11.3\ \mu\text{rad}$ and $\sigma_{\mathrm{V}} = 5.9\ \mu\text{rad}$ during the experiments described in the work of Marx *et al.*[10] This results, according to Eq. (7), in a minimum possible polarization purity of 1.6×10^{-10} which agrees well with the measured value of $2.4(9) \times 10^{-10}$ at a photon energy of 6.456 keV, at least if one considers the uncertainty of the stated values and the experimental error bar. The usage of collimating lenses would not have had a large influence on the horizontal divergence because of the horizontal source size. In the vertical direction, an almost parallel beam would be possible. The current upgrade of the ESRF will improve the horizontal extension of the electron beam dramatically and, thus, the possibility of collimation. Theoretically, a reduction in the divergence to about 1 μrad will be possible with the new storage ring leading to purities in the order of 10^{-12}. Another possibility to decrease the divergence is the use of asymmetrically cut crystals in front of the polarimeter. A reduction by a factor of 3. . .5 is feasible. At the same time, the beam will be expanded by the same factor. Depending on the spatial acceptance of the channel-cut crystals, an improvement of the polarization purity by about one order of magnitude is possible with only minor losses in flux.

Fundamental limitations of the polarization purity

(a) Multiple wave diffraction





σ → π polarization transfer limits the purity for multiple reflections

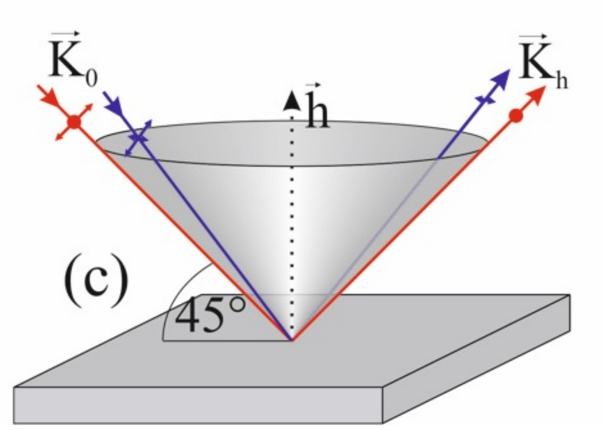
(b) Beam divergence

For a Gaussian beam with H/V divergencies σ_H and σ_V:

$$\boxed{\delta \approx \sigma_{\mathrm{H}}^2 + \sigma_{\mathrm{V}}^2}$$

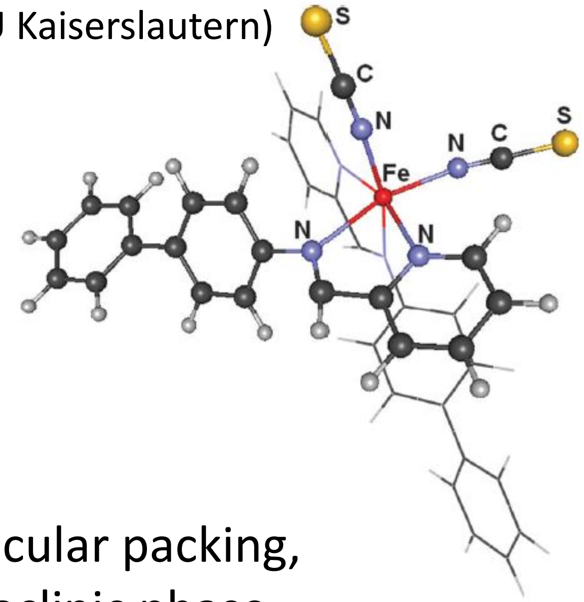
Polarization purity

$$\boxed{\delta \sim 10^{-12} \text{ for } \sigma_V, \sigma_H \sim 1\ \mu\text{rad}}$$



Charge anisotropy in a iron spin crossover (SCO) compound

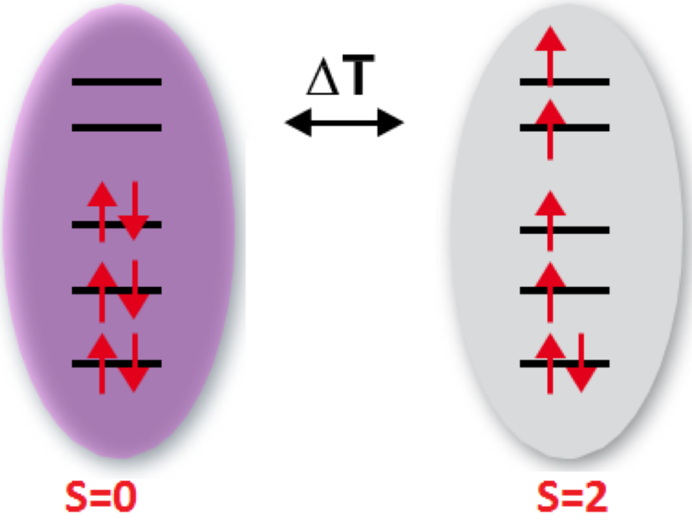
Collaboration with L. Scherthan, J. Wolny, V. Schünemann (TU Kaiserslautern)



$[Fe(PM\text{-}BiA)_2(NCS)_2]$

SCO complex with a strong EFG at the position of the Fe atom

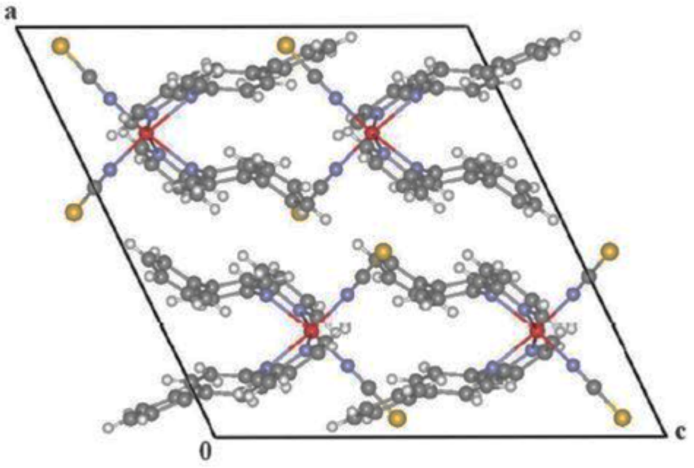
T > 170 K



Low-spin

High-spin

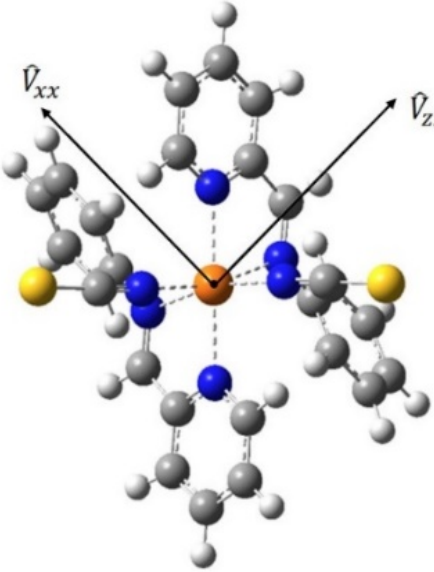
Molecular packing, monoclinic phase



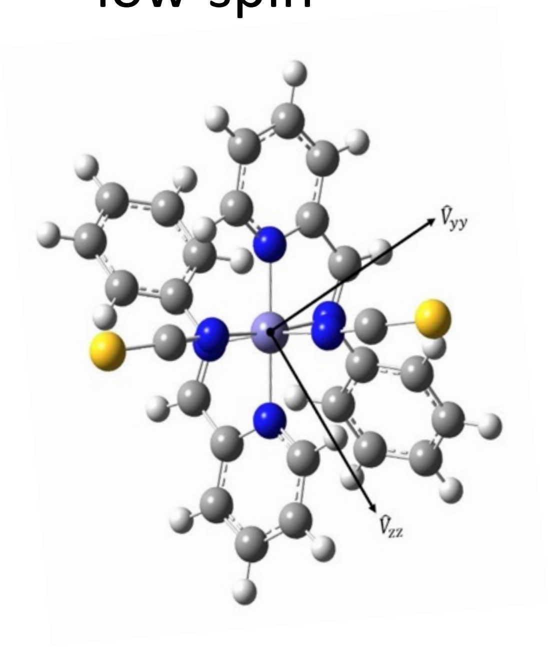
Change of EFG orientation upon spin-crossover

results of DFT calculations **$[\mathrm{Fe(PM\text{-}BiA)_2(NCS)_2}]$**

high-spin

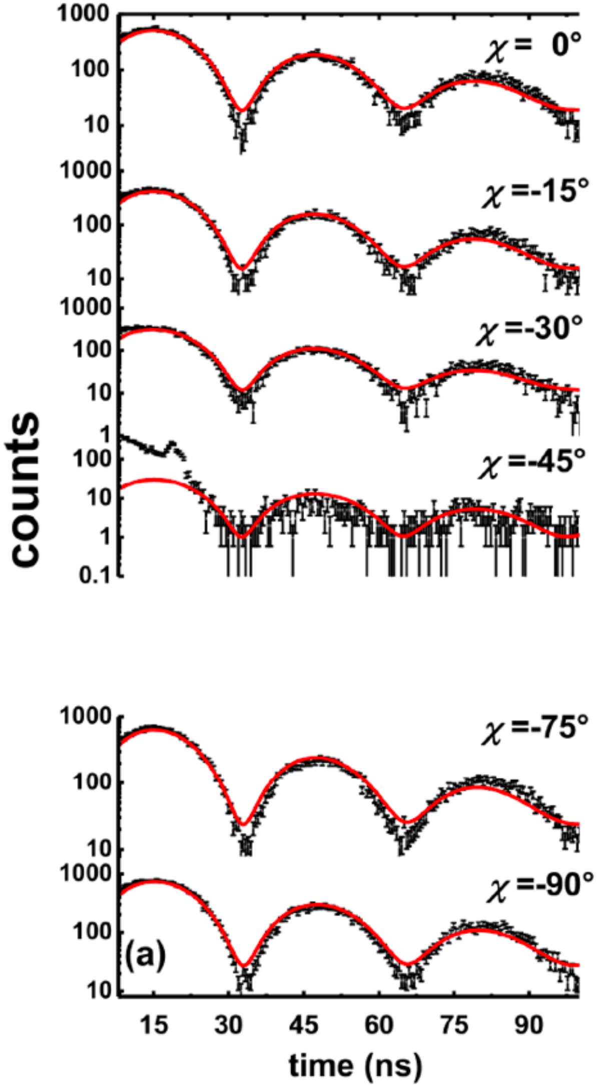


low-spin

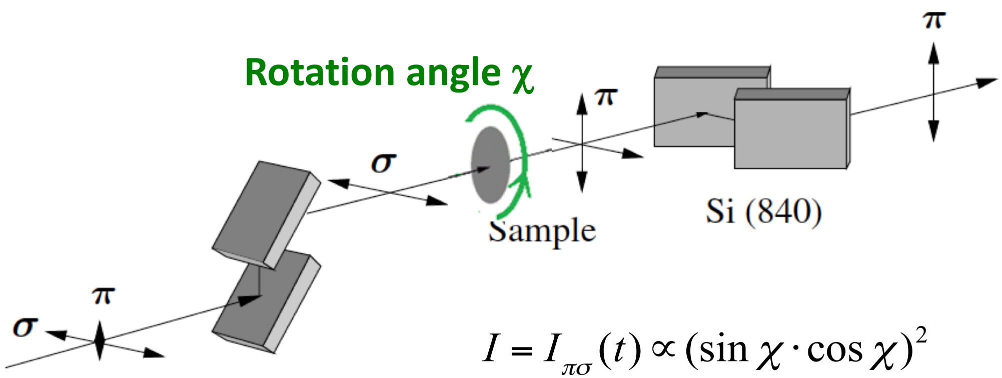


Nuclear resonance polarimetry of an SCO complex

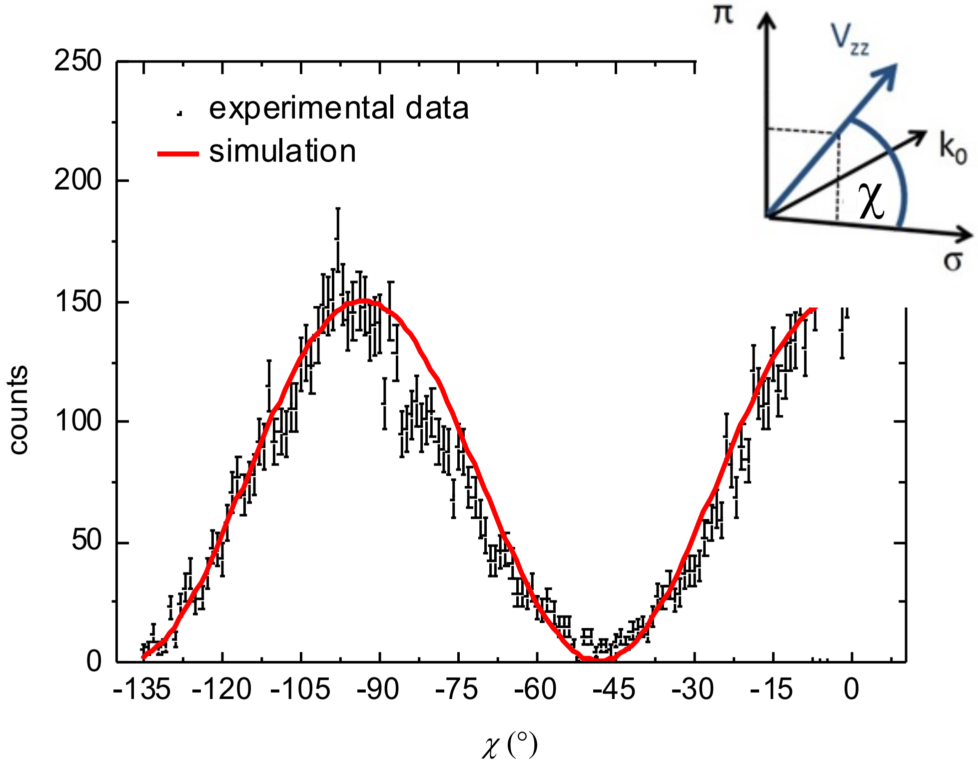
T = 220 K, high-spin



Rotation angle χ



$$I = I_{\pi\sigma}(t) \propto (\sin\chi \cdot \cos\chi)^2$$



Nuclear resonance polarimetry of an SCO complex

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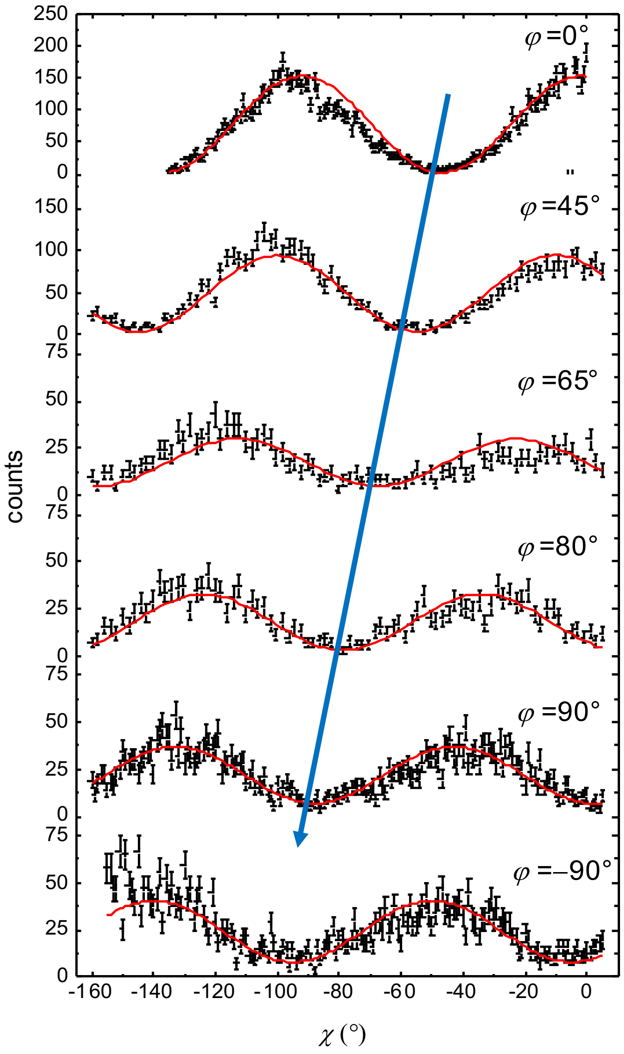
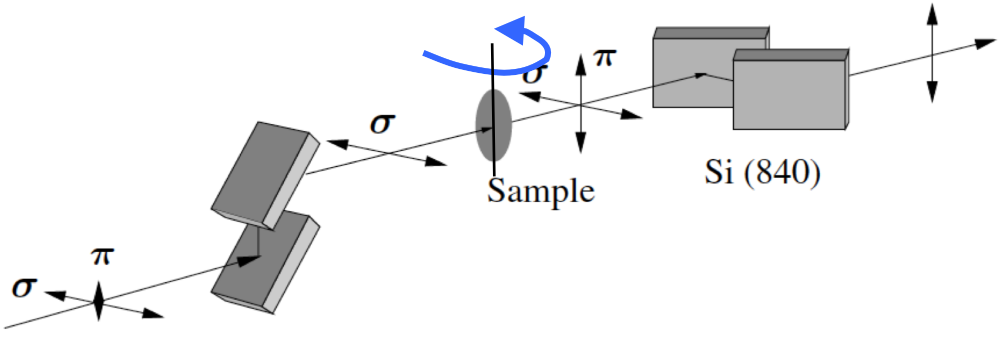
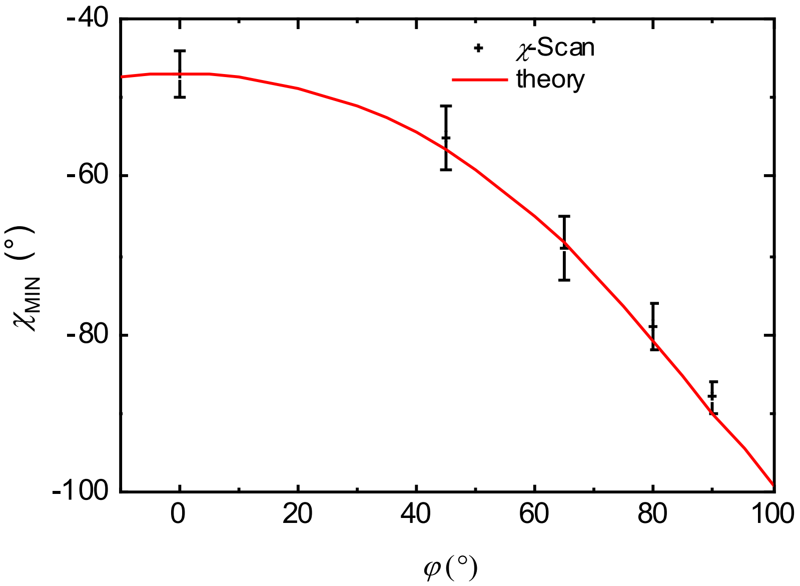
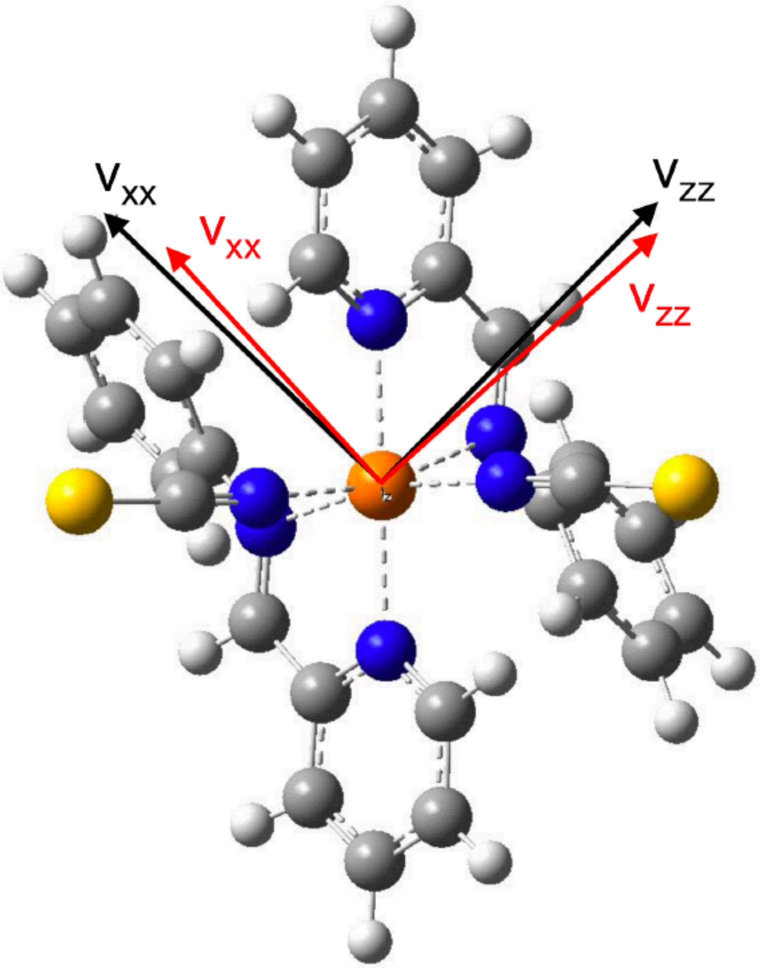


Rotation angle φ





Results: Electric field gradient in the high-spin state

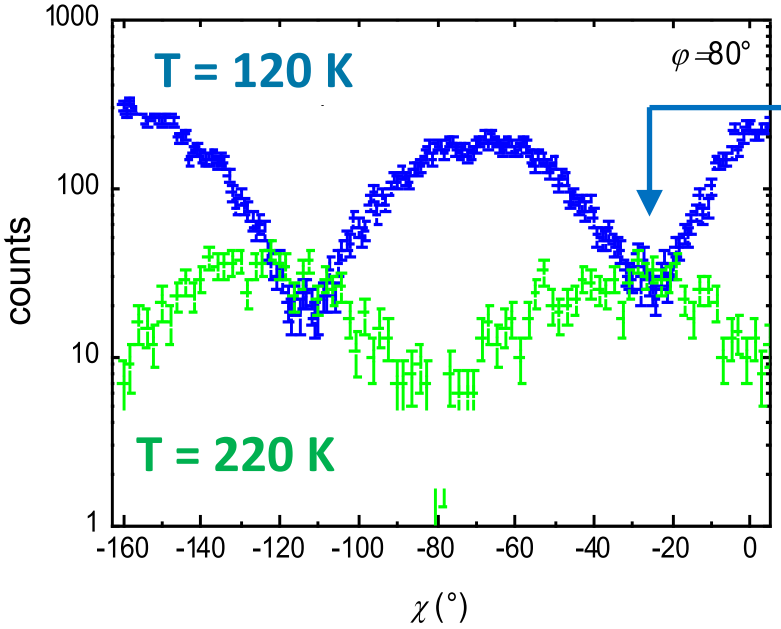


DFT calculations

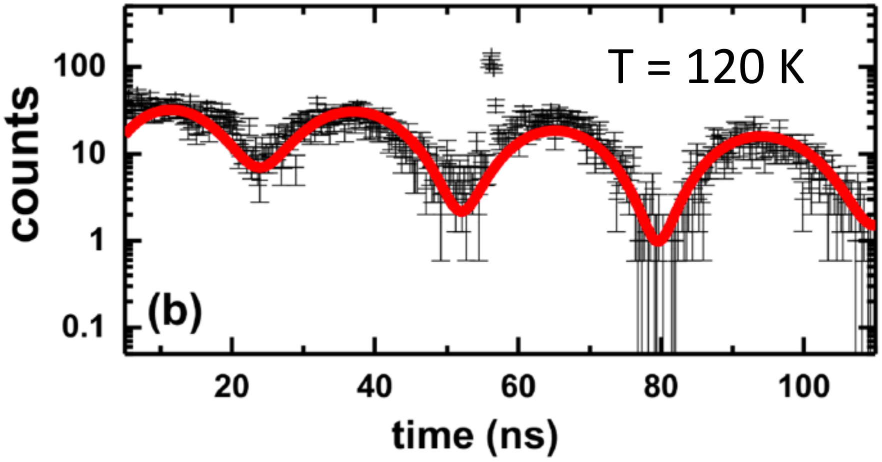
Measured data

Nuclear resonance polarimetry of an SCO complex

Experimental results: Temperature dependence



Significant contribution from high-spin phase

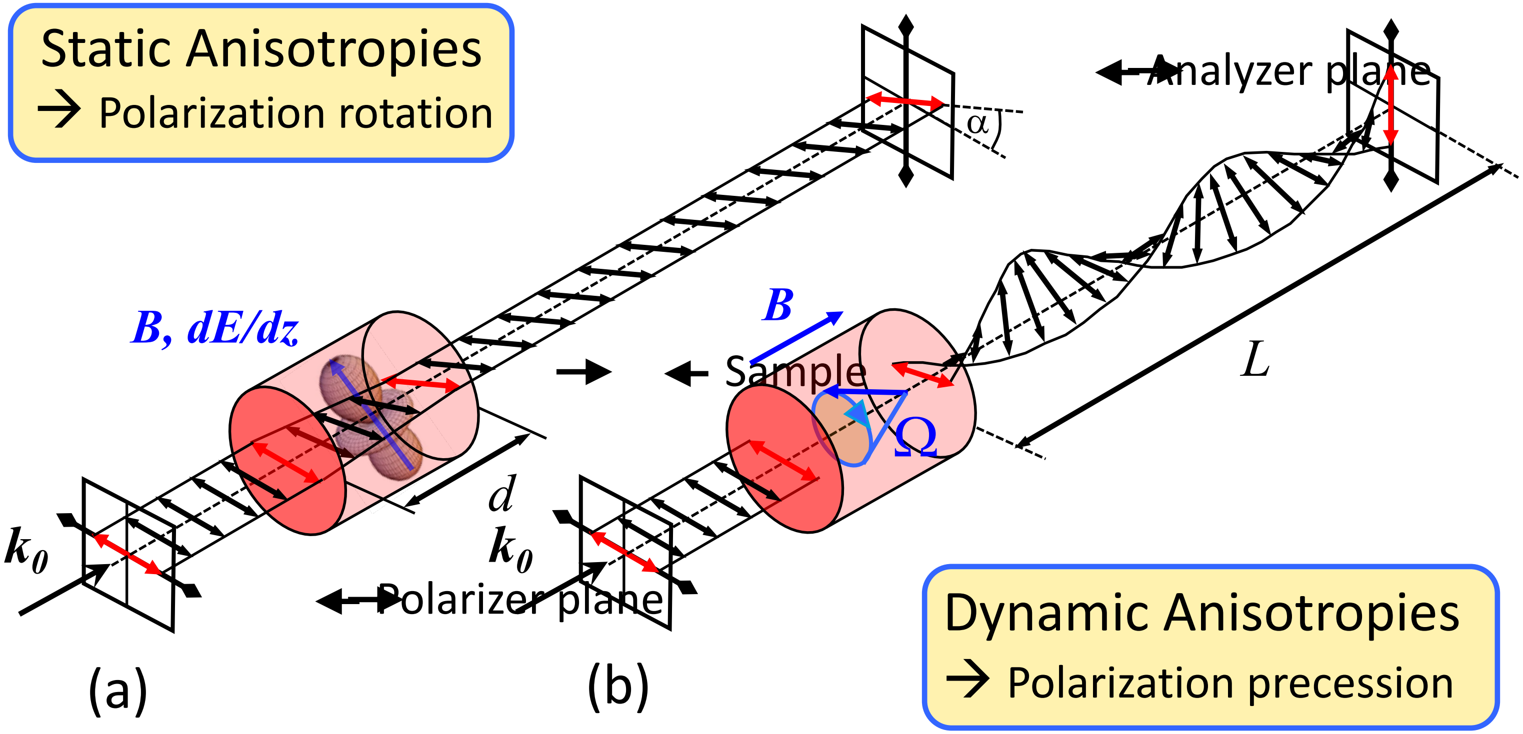


→ Change of EFG orientation due to the SCO

Evaluation reveals:
78 % low-spin, 22 % high-spin
(oriented !)

Diploma thesis Lena Scherthan, Uni Kaiserslautern (2016)

Probing Anisotropies in Condensed Matter via Polarimetry



Static Anisotropies
→ Polarization rotation

Dynamic Anisotropies
→ Polarization precession

Probing charge anisotropies in correlated materials

→ Adressing selected orbitals via resonant x-rays

Probing spin excitations in magnetic materials

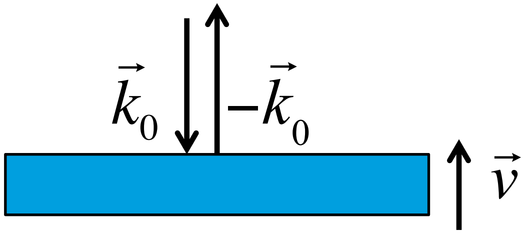
→ PRL 112, 117205 (2014)

Energy Transfer in Inelastic Light Scattering

Linear Doppler effect:

$$\Delta E = \Delta\vec{k} \cdot \vec{v}$$

$$\Delta\vec{k} = 2\vec{k}_0$$

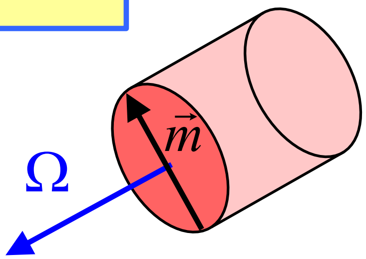


→ scattering of light from a **moving object with** broken **translational invariance**

→ solid-state excitation: **Phonon**
(corresponds to a moving mirror)

Angular Doppler effect:

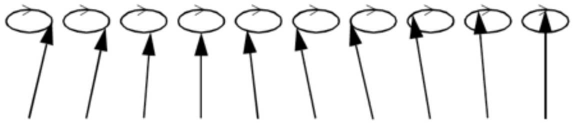
$$\Delta E = \Delta\vec{L} \cdot \vec{\Omega}$$



→ scattering of light from a **rotating object with** broken **rotational invariance**
(optical anisotropy)

A half-wave plate acts an angular mirror → $\Delta L = 2\hbar$

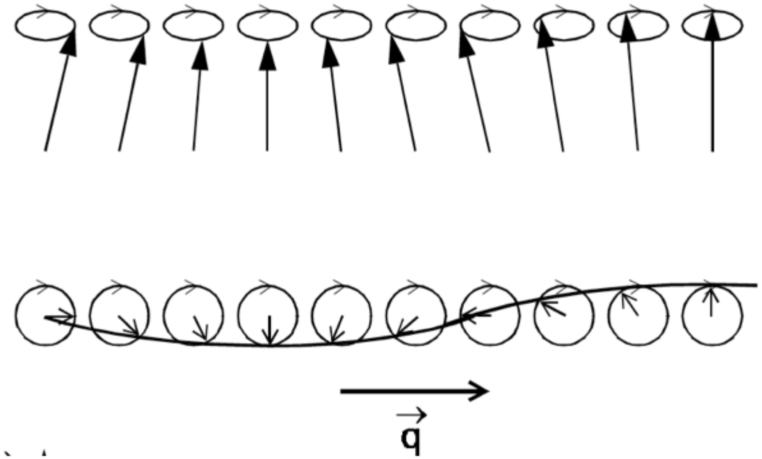
→ solid-state excitation: **Magnon**
(corresponds to a rotating phase retarder)

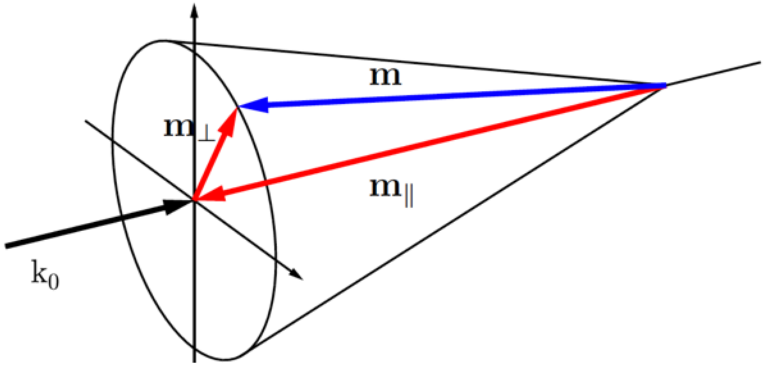


Magnons (Spinwaves)

Rotational anisotropy in solids is generated by magnetization

Dynamic anisotropy in spin waves:





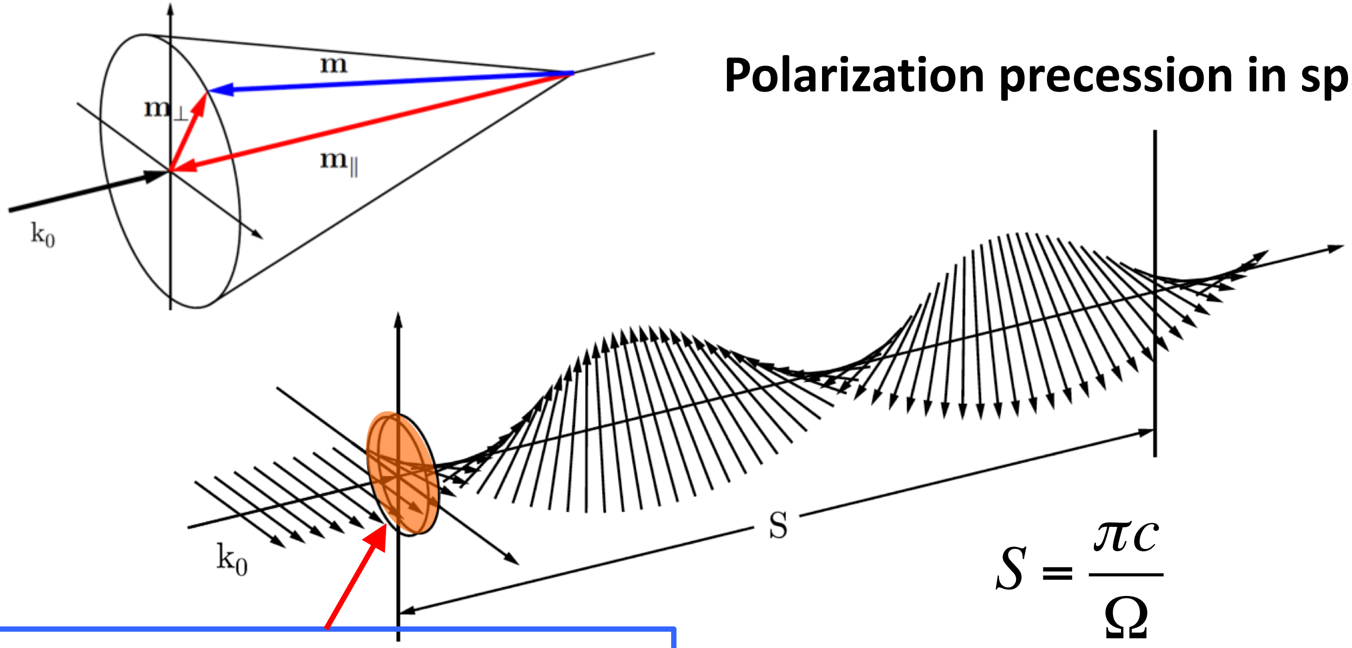
Resonant Scattering from Magnons: Twisted Polarization

$$\left[A_S\right]_{circ} = i\,e^{i\,2\Omega t}A_- + e^{-i2\Omega t}A_+$$

Transmission through a rotating half-wave plate / Light scattering from a magnon

Transformation into linear basis:

$$\left[A_S\right]_{lin} = \cos 2\Omega t\; A_H - \sin 2\Omega t\; A_V$$

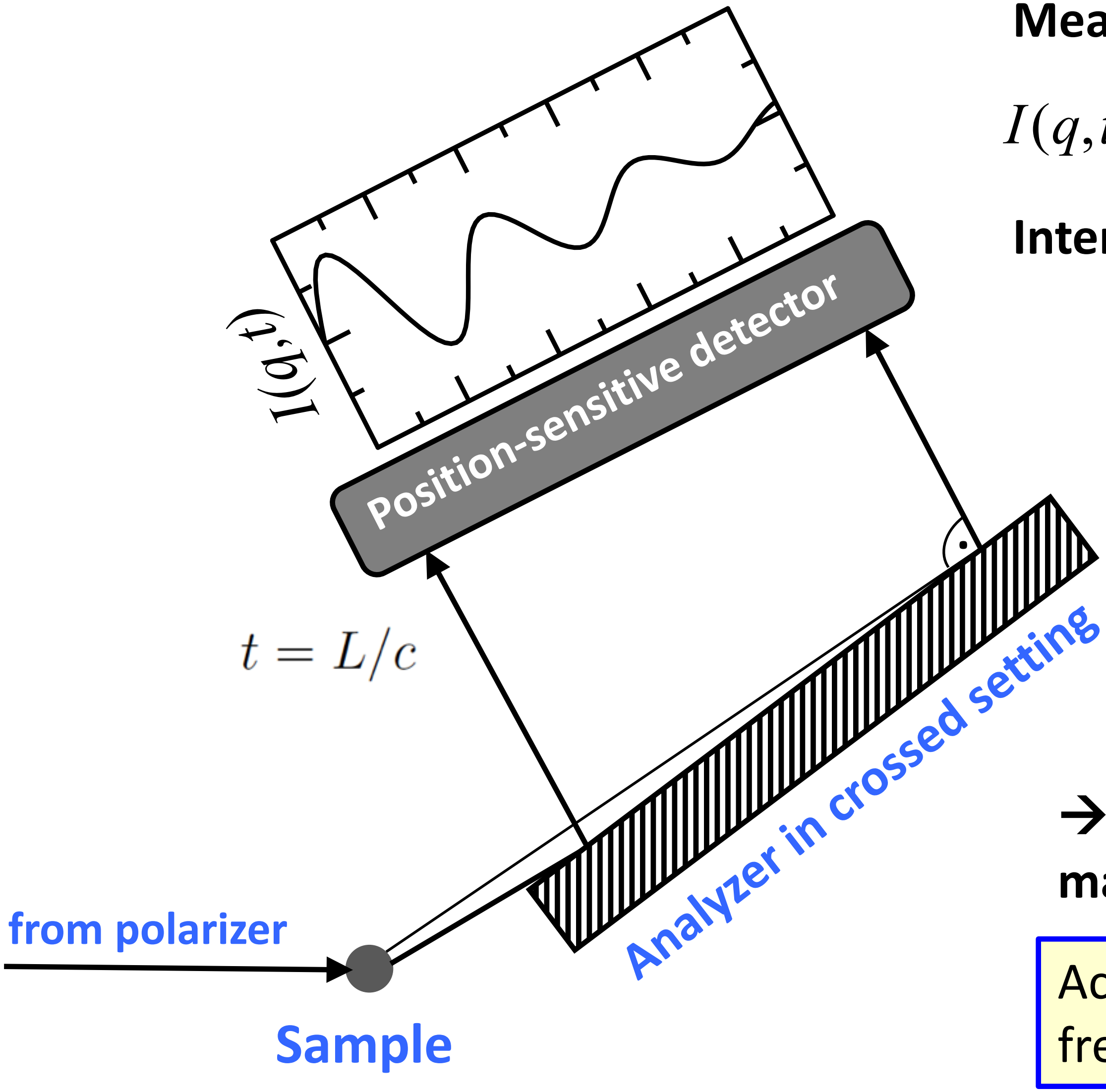


Polarization precession in space

$$S = \frac{\pi c}{\Omega}$$

Sample with magnon excitation

Resonant Scattering from Magnons: Polarization Analysis



Measured signal:

$$I(q,t) = I_B + I_0 \int_{-\infty}^{\infty} S(q,\Omega)\cos 2\Omega t \, d\Omega$$

Intermediate scattering function

- Compare with Neutron Spin Echo
- Energy transfer encoded in polarization
- **Independent of energy bandwidth !**

→ Probing low-energy magnetic excitations

Accessible range of spinwave frequencies: 1 – 500 GHz

Outlook: A synchrotron Mössbauer source for ^{40}K

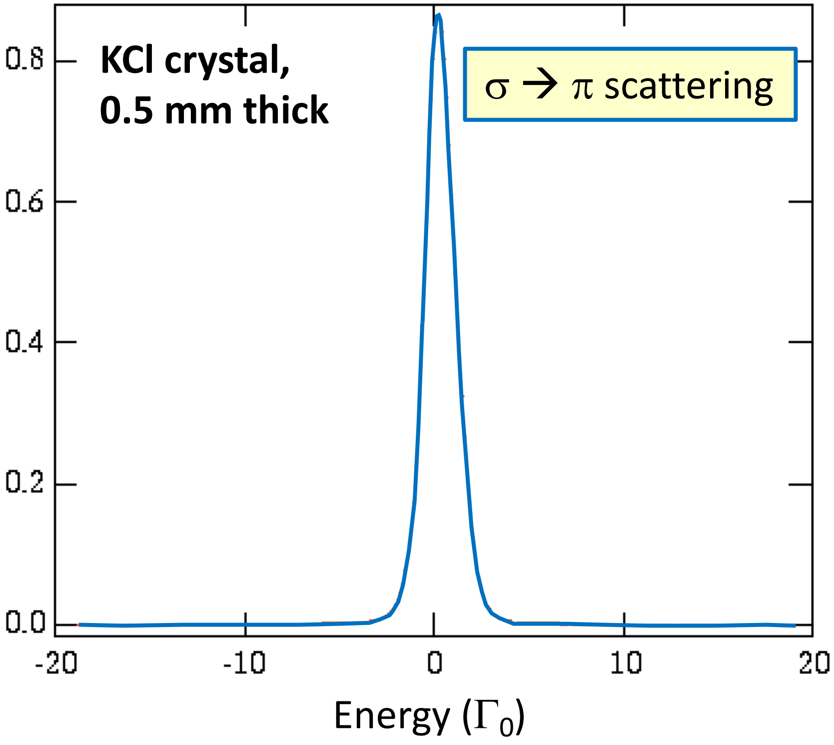
Transition energy: 29.8 keV
Lifetime: 8 ns
Natural abundance: 0.01 %

Relevant for:

- **biological functions**
- **unconventional superconductors**

Requires extension of polarimetry to high energies !

6 T magnetic field in Faraday geometry

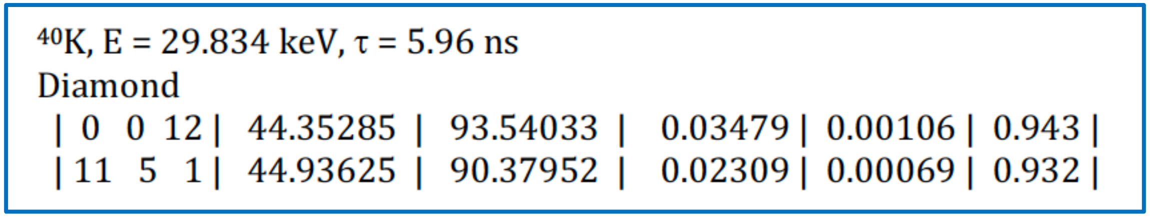


KCl crystal, 0.5 mm thick

$\sigma \rightarrow \pi$ scattering

BMBF
Verbund
projekt
FSU Jena

Potential reflections:



^{40}K, E = 29.834 keV, τ = 5.96 ns
Diamond

0 0 12	44.35285	93.54033	0.03479	0.00106	0.943
11 5 1	44.93625	90.37952	0.02309	0.00069	0.932